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Temperature dependence of magnetoelastic properties of Fe_{100-x}Si_x (5 < x < 20)

Abstract

Tetragonal magnetostriction (λ^2) and elastic constants (c' , c_{44} , and c_{11}) for Fe_{100-x}Si_x were measured as a function of temperature (T). Compositions corresponding to the disordered A2 ($x = 5$), ordered D0₃ ($x = 19.8$), and mixed ($x = 11.6$) phases, were investigated. The magnetoelastic coupling ($-b_1$) was determined for $77 < T < 300$ K and compared with those of Fe-Ga, Fe-Ge, and Fe-Al. Both $\lambda^2(T)$ and $-b_1(T)$ of Fe-Si behave similarly to those of Fe-Ge, while other notable differences exist between the measured properties of Fe-Si and those of the other three alloys. Due to the early establishment of short range order, Fe-Si exhibits a positive, although small, slope in $\lambda^2(T)$ at 5 at. % Si, and a remarkable drop in $-b_1$ before the solubility limit. The weaker softening of the tetragonal shear modulus with the addition of Si and the lack of strong anharmonic effects in the Fe-Si lattice inferred from the weak T -dependence of all the moduli suggest that Fe-Si exhibits more structural stability than the other three alloys. The distinctive behavior is likely due to the smaller size of Si compared to the sizes of Ga, Ge and Al, and therefore to the effect of the larger size difference between Fe and Si in the Fe-Si lattice.

Keywords

association, elastic constants, iron alloys, magnetoelastic effects, magnetostriction, shear modulus, short-range order, silicon alloys, solubility

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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Temperature dependence of magnetoelastic properties of $\text{Fe}_{100-x}\text{Si}_x$ ($5 < x < 20$)

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Tetragonal magnetostriction ($\lambda^{T,2}$) and elastic constants (c' , c_{44} , and c_{11}) for $\text{Fe}_{100-x}\text{Si}_x$ were measured as a function of temperature (T). Compositions corresponding to the disordered A2 ($x=5$), ordered D0_3 ($x=19.8$), and mixed ($x=11.6$) phases, were investigated. The magnetoelastic coupling ($-b_1$) was determined for $77 < T < 300$ K and compared with those of Fe-Ga, Fe-Ge, and Fe-Al. Both $\lambda^{T,2}(T)$ and $-b_1(T)$ of Fe-Si behave similarly to those of Fe-Ge, while other notable differences exist between the measured properties of Fe-Si and those of the other three alloys. Due to the early establishment of short range order, Fe-Si exhibits a positive, although small, slope in $\lambda^{T,2}(T)$ at 5 at. % Si, and a remarkable drop in $-b_1$ before the solubility limit. The weaker softening of the tetragonal shear modulus with the addition of Si and the lack of strong anharmonic effects in the Fe-Si lattice inferred from the weak T -dependence of all the moduli suggest that Fe-Si exhibits more structural stability than the other three alloys. The distinctive behavior is likely due to the smaller size of Si compared to the sizes of Ga, Ge and Al, and therefore to the effect of the larger size difference between Fe and Si in the Fe-Si lattice.

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I. INTRODUCTION

The search for a complete understanding of the mechanism behind the high magnetostriction of Fe-Ga and the unusual dependence of the magnetostriction on Ga concentration, led to the exploration of other Fe-based alloys. It became clear that the itinerant aspect of the $3d$ electrons, actively involved in the magnetism of iron, is a strong source of variability in the magnetoelastic behavior of the alloys. Studies of Fe-alloys where the solute element is in a particular location with respect to Ga in the periodic table has been of recent interest. The elements involved are similar in some ways, as well as different in others: full (Ga, Ge) and empty (Al, Si) d -shell elements or Group III (Al, Ga) and Group IV (Si, Ge) elements. The purpose of engaging this block of elements is to trace the influence of size and valence-electron number of the solute on the magnetoelastic coupling of the alloy. The magnetoelastic properties of Fe-Ga,^{1,2} Fe-Ge,^{1,3} and Fe-Al^{1,4} have been measured as a function of temperature; theoretical calculations (at 0 K) exist for Fe-Ga⁵ and Fe-Ge.^{3,6} Fe-Si in bulk has been the subject of two recent studies: one experimental, measuring the tetragonal magnetostriction dependence on composition, structure, and thermal treatment at room temperature,⁷ and one theoretical, determining magnetic anisotropy coefficients and magnetostriction at absolute zero.⁸ This article presents results of an

extended experimental investigation of bulk $\text{Fe}_{100-x}\text{Si}_x$ ($5 < x < 20$), which include: (i) temperature dependence of the tetragonal magnetostriction, $\lambda^{T,2} = (3/2)\lambda_{100}$, for $77 < T < 300$ K, (ii) temperature dependence of the shear moduli $c' = (c_{11} - c_{12})/2$ and c_{44} (tetragonal and rhombohedral, respectively) and of the longitudinal modulus, c_{11} , for $7 < T < 300$ K, (iii) temperature dependence of the magnetoelastic constant $-b_1$ for $77 < T < 300$ K. The results allow for comparisons of magnetoelastic properties between the four alloys described above (Fe- X , with $X = \text{Al, Si, Ga}$ and Ge) as a function of temperature, completing the above-mentioned four-element study. The results also make the comparison between the experimental and theoretical results for Fe-Si possible over a lower temperature difference (77 K to 0 K rather than 300 K to 0 K).

II. EXPERIMENT

Three compositions, corresponding to the disordered A2, ordered D0_3 , and mixed phase regions that are characteristic to $\text{Fe}_{100-x}\text{Si}_x$ in different ranges of the $0 < x < 25$ interval,⁹ were chosen for the study. Single crystal ingots of Fe-Si were prepared using the Bridgman method.² All samples were slow cooled at 10 K/min after post-growth annealing at 1273 K. The composition was measured to be $x = 5, 11.6$, and 19.8 at. % Si (± 0.3 at. %), by electron probe microanalysis using pure iron and silicon standards for calibration. The phases were confirmed to be A2, mixed, and D0_3 , in the order of increasing Si content, by XRD. At each composition, a rectangular

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parallelepiped (sides between 1.2 and 3.3 mm) was cut and polished with its planar surfaces parallel to {100} crystallographic planes of the Fe sublattice. Disk samples (2.5 mm thickness and 6.35 mm diameter) at matching compositions that were previously fabricated for the room-temperature study were used for the low-temperature magnetostriction measurements. The standard rotating-sample technique,³ at temperatures between 77 K and 300 K, in a 20 kOe field (above saturation), was used to measure the tetragonal magnetostriction values. Measurements of the three independent elastic moduli of the alloys (all samples are cubic) were done with resonant ultrasound spectroscopy (RUS) on the rectangular parallelepipeds, at temperatures between 7 K and 300 K. A magnetic field of 20 kOe was applied along the longest side of the parallelepiped during the measurements.

The RUS samples were used in their original as-grown slow cooled state (SC). The disk samples, however, had been quenched (Q) for the previous room-temperature study.⁷ That study showed that if the alloy concentration belongs to one of the pure-phase regions, either disordered ($0 < x < 10$) or ordered ($15 < x < 25$), heat treatment has no effect on the structure, and therefore on the magnetostriction, of Fe-Si. For that reason, the 5 and 19.8 at. % Si single-phase disk samples were left in their quenched state. On the other hand, quenching was shown to have an effect on mixed-phase samples and the 11.6 at. % Si disk sample was reheated and slow cooled at 10 deg/min to make it compatible with the similar RUS sample.

III. RESULTS AND DISCUSSION

The saturation-value tetragonal magnetostriction, $\lambda^{y,2}$, for the three investigated Fe-Si crystals is shown in Fig. 1 as a function of temperature. Previously reported relevant data are included in the figure: room temperature values measured on the same samples in both SC and Q states,⁷ and theoretical values at 0 K⁸ at Si concentrations closest to the experimental ones. A phase-specific temperature dependence, similar to that seen in Fe-Ge, is found for Fe-Si. Both Group-

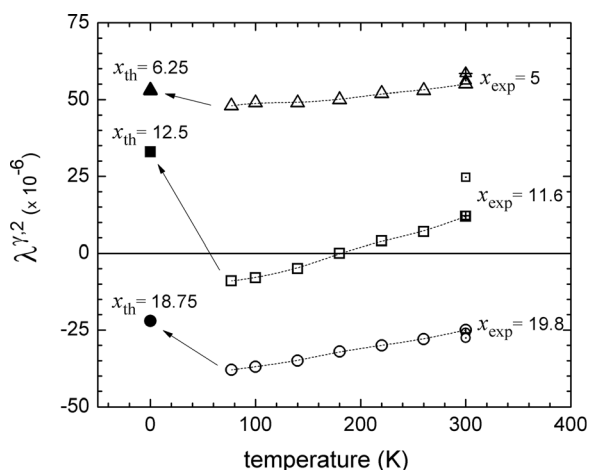


FIG. 1. Temperature dependence of the tetragonal magnetostriction of the three $\text{Fe}_{100-x}\text{Si}_x$ samples studied (clear symbols); 300 K data from Ref. 7 is added, in both slow cooled (crossed symbols) and quenched (dotted symbols) states. The filled symbols represent 0 K theoretical results⁸ at the compositions closest to the experimental ones.

IV solute-element alloys are known to have stable D0_3 ordered structures. The expected behavior of a continuously decreasing $|\lambda^{y,2}|$ as temperature increases is found only for the D0_3 phase, at $x = 19.8$ at. % Si. The opposite is found for the mixed phase, behavior that has been explained for Fe-Ge through a simple rule of mixtures between the characteristic $\lambda^{y,2}(T)$ curves of the A2 and D0_3 phases present in the sample. This qualitative explanation applies to Fe-Si as well, however, with one difference. Short range order (SRO) of both the B2 and D0_3 type is likely to occur in $\text{Fe}_{100-x}\text{Si}_x$ at $5 < x < 8$ at. %, (estimated length scale of 3.4 nm),⁷ in accord with the phase diagram⁹ and structure measurements. While the characteristic $\lambda^{y,2}(T)$ for D0_3 is found from the 19.8 at. % Si sample, a curve for pure B2 cannot be measured. If the features of $\lambda^{y,2}$ for B2 are not entirely different than those of D0_3 (i.e. negative and increasing $\lambda^{y,2}$ values), the rule of mixtures between the three present phases explains the positive $d\lambda^{y,2}/dT$ for the $x = 11.6$ sample. The slight increase in $\lambda^{y,2}$ versus T for the 5 at. % Si sample is consistent with the early presence of SRO (B2 and/or D0_3) in Fe-Si. The significant volume fraction of SRO in Fe-Si at concentrations lower than the solubility limit has been associated with the earlier-than-expected occurrence of the first peak in $|b_1(T)|$, when compared to the other three alloys in the block, as a function of e/a (number of valence electrons per atom in a given volume).¹⁰ One proposed explanation¹ for this atypical behavior of Fe-Si is the smaller size of Si, compared with Ga, Ge, and Al.

The three independent elastic constants c' , c_{44} and c_{11} , of Fe-Si at the compositions studied are shown in Fig. 2 as a function of temperature. The error in the measurement increases with the value of the constant (0.2, 0.6, and 3.5% for c' , c_{44} , and c_{11} , respectively). Several notable behaviors are observed, when contrasted with the other Fe-X alloys considered. The dramatic softening of c' , as x increases between the values that correspond to the A2 and the D0_3 phases, does not occur in Fe-Si. To compare, c' for D0_3 Fe-Si is 75% of the pure Fe ($x = 0$) c' value, while for Fe-Ga the corresponding value is 16%. For Fe-Ge and Fe-Al, the same ratio is $\sim 40\%$ (300 K values are used in all estimates). Moreover, $c'(x)$ for Fe-Si is not linear through the solubility limit, as in the case of the other three alloys. The weak temperature dependence of all the constants of Fe-Si indicates a normal elastic behavior with low anharmonic effects at all three compositions.

Combining the magnetostriction and elasticity data, the tetragonal magnetoelastic coupling constant, $-b_1 = 2\lambda^{y,2}c'$, was found, as shown in Fig. 3. Interpolations for the measured tetragonal magnetostriction (order 2 polynomial fits) were used to find $\lambda^{y,2}(T)$ at the same temperatures where c' was measured in the $77 < T < 300$ K interval. A temperature dependence similar with that of $\lambda^{y,2}$ is seen for the $-b_1$ curves at all three compositions.

Furthermore, in Fig. 4, $-b_1$ of Fe-Si is plotted together with the corresponding curves of the companion Fe-X alloys at 77 K (e/a is the universal scaling parameter introduced above). As in the 300 K case, the Fe-Si results stand out with an early drop in $-b_1$. For the other three alloys, a large drop in $-b_1$ occurs only after the composition reaches the

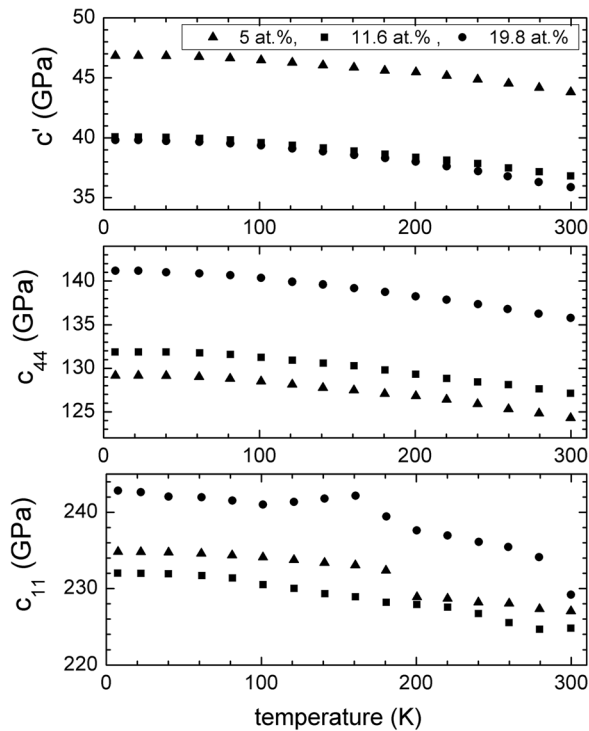


FIG. 2. Temperature dependence of the elastic constants of the three Fe-Si samples studied (all cubic symmetry), in 20 kOe magnetic field.

solubility limit, at a common value of $e/a \approx 1.34$. This observation concurs with the effects of the particular size of the Si atom. Si, the smallest in the group of four relative to Fe, is expected to induce more stability to the bcc structure of the alloy and an early appearance of ordered phases. The higher structural stability in Fe-Si is also implied by the weaker softening of the tetragonal shear constant, c' , versus x .

IV. SUMMARY

The temperature dependence of the tetragonal magnetostriction (for $77 < T < 300$ K) and the elastic constants (for $7 < T < 300$ K) were measured for $\text{Fe}_{100-x}\text{Si}_x$ at $x = 5, 11.6$, and 19.8. The three compositions correspond to the charac-

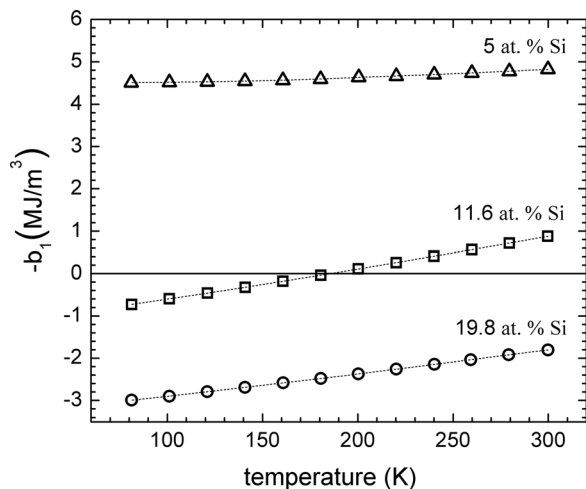


FIG. 3. Temperature dependence of the tetragonal magnetoelastic coupling, $-b_1 = 2\lambda\gamma^2 c'$, of Fe-Si at the three compositions studied.

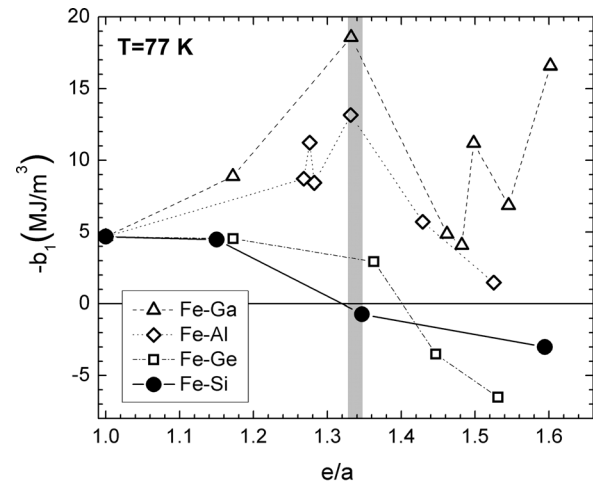


FIG. 4. Tetragonal magnetoelastic coupling, $-b_1$, at $T = 77$ K, for Fe-Si and three other alloys versus e/a (the solubility limit is marked in gray).

teristic structures of Fe-Si in the $0 < x < 25$ range. The measurements allow the calculation of the tetragonal magnetoelastic coupling constant $-b_1$ for $77 < T < 300$ K. Notable differences between the obtained properties of Fe-Si and those of Fe-Ga, Fe-Ge, and Fe-Al, previously determined, are (i) a slight positive slope in $\lambda\gamma^2$ at 5 at. % Si and a significant drop in $-b_1$ before the solubility limit, both consistent with the early presence of SRO in Fe-Si, (ii) a weaker c' softening with increasing amount of solute, which is concurrent with a higher structure stability induced by Si and (iii) a lack of strong anharmonic effects in the Fe-Si lattice at any of the three studied compositions.

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